Magnetic percolation effect on the spontaneous Hall resistivity and magnetoresistance of $La_{1-x}A_xCoO_3$ ($A = Ca, Sr$; $0.1 \le x \le 0.5$)

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Abstract

The Hall resistivity and magnetoresistance of $La_{1-x}A_xCoO_3$ (A = Ca, Sr) are investigated. The spontaneous Hall coefficient reaches maximum near the Curie temperature for each doping level, and achieves the largest magnitude near the magnetic percolation threshold. The physical significance of these results are discussed.

The Hall resistivity ρ_{xy} of a metallic system with localized magnetic moments is generally given by $\rho_{xy} = R_0 B + R_s(\mu_0 M)$, where $R_0 = 1/(ne)$ is the Hall coefficient for a conducting carrier density n and charge e, B is the magnetic induction, and R_s the anomalous Hall coefficient associated with the magnetization M of a sample. Conventional theory attributes a finite R_s to asymmetric spin-orbit scattering of carriers, and predicts the relations between ρ_{xy} and the longitudinal resistivity (ρ_{xx}) as either ($\rho_{xy} \propto \rho_{xx}$) for the *skew scattering* mechanism [1], or ($\rho_{xy} \propto \rho_{xx}^2$) for the *side-jump* mechanism [2]. However, our recent studies of ferromagnetic cobaltites $La_{1-x}Ca_xCoO_3$ ($0.2 \le x \le 0.5$) [3] reveal novel properties and a record value of $R_s(\approx 1.4 \times 10^{-6}\ m^3/C)$ in $La_{0.8}Ca_{0.2}CoO_3$ that cannot be explained by conventional theory. We have attributed these results to the existence of multiple spin configurations of trivalent Co-ions, (Co^{3+}: $t_{2g}^4 e_{2g}^2$, $S = 2$; Co^{III}: $t_{2g}^6 e_{2g}^0$, $S = 0$), strong spin fluctuations near T_{Curie}, and magnetic percolating effect as a function of temperature (T) and doping level x. In this work, we extend our studies to $La_{1-x}Sr_xCoO_3$ ($x = 0.2, 0.5$), and compare the properties of Sr-doped with Ca-doped systems, and also with ferromagnetic $La_{1-x}A_xMnO_3$.

The substitution of divalent Ca or Sr in $LaCoO_3$ results in tetravalent Co^{4+} ($t_{2g}^3 e_{2g}^2$) ions, which stabilizes the high-spin Co^{3+} ions, and ferromagnetism is established for $x > 0.18$ [3]. For a given x, the fraction of high-spin Co^{3+} among the trivalent Co-ions increases with T, and reaches $\sim 50\%$ at $T \approx 110$ K. Interestingly, $T_{Curie} \approx 110$ K in $La_{0.8}Ca_{0.2}CoO_3$, and $R_s(T \approx T_{Curie})$ reaches a record value among all known stochiometric ferromagnetic materials [3]. The enhancement of R_s near the magnetic percolation threshold ($x = 0.2$) may be understood as follows: The spontaneous Hall effect is proportional to the spin-orbit coupling strength λ_{so}, and $\lambda_{so} \sim [(\mathbf{k} \times \sigma) \cdot \nabla V_c]$, where V_c is the crystalline potential. Ferromag-

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netic cobaltites consist of high-spin hole-rich clusters embedded in a background of low-spin hole-poor matrix. Therefore, a maximum magnitude in ∇V_c is expected near the magnetic percolation threshold, yielding enhanced spin-orbit scattering and large R_s. This argument is consistent with the experimental observation in ferromagnetic $La_{1-x}Ca_xCoO_3$, as summarized in Figures 1(a)-(b). Similar behavior is also observed in ferromagnetic $La_{1-x}Sr_xCoO_3$, as shown in Figures 1(c)–(d). Both systems exhibit maximum ρ_{xy} and R_s for $x = 0.2$. In addition, $R_s(T)$ reaches maximum at $T <\sim T_{Curie}$, suggesting the relevance of spin fluctuations. Moreover, except for $La_{0.8}Ca_{0.2}CoO_3$, ρ_{xx} of the cobaltites is reduced under an external magnetic field H, and the magnitude of magnetoresistance, $\Delta R_H \equiv [\rho_{xx}(H) - \rho_{xx}(0)]/\rho_{xx}(0)$, reaches maximum near T_{Curie}, as shown in Figures 1(a) and 1(c). These data are consistent with the suppression of disorder spin scattering under a finite H. On the other hand, $\Delta R_H(T)$ changes sign near maximum $R_s(T)$ for $La_{0.8}Ca_{0.2}CoO_3$. This anomalous behavior is not yet understood.

For a given doping level x, the magnitude of R_s in $La_{1-x}Sr_xCoO_3$ is much smaller than that in $La_{1-x}Ca_xCoO_3$, possibly due to stronger spin fluctuations in the latter, whose Curie temperatures are lower so that more low-spin Co^{III} ions may switch to the high spin states near T_{Curie}. Another possibility is that carriers moving in a non-trivial spin background may acquire a "Berry phase" [4] that affects the motion of carriers in the same way as does an external magnetic field. Recent theory based on the concept of Berry phase [5] has shown that for $T < T_{Curie}$, $(R_s/R_0) \propto \exp[-E_c/(k_B T)]$, where E_c is the "core energy" for creating topologically non-trivial spin configurations, and that $E_c \propto T_{Curie}$. The relevance of Berry phase is consistent with our experimental observation that R_s in $La_{1-x}Sr_xCoO_3$ is significantly *smaller* than that in $La_{1-x}Ca_xCoO_3$ for a given x, because T_{Curie} and E_c of Sr-doped cobaltites are larger than those of the Ca-doped cobaltites. However, the Berry phase theory for spontaneous Hall effect has been developed for $La_{1-x}A_xMnO_3$, where the Hund's on-site exchange interaction energy (J_H) is much larger than the hopping energy (t) of the carriers. Although this assumption may be relaxed, several differences are noteworthy: R_s in cobaltites is significantly *larger* than that in the manganites, whereas $|\Delta R_H|$ in the cobaltites is several orders of magnitude *smaller*; R_0 and R_s are of the same sign in the cobaltites, and are opposite in the manganites; the conducting t_{2g} electrons of the cobaltites move in a background of core electrons ($t_{2g}^3 e_g^2$) of opposite spin orientation, whereas the spins of conducting e_g electrons in manganites are parallel to those of the core electrons ($t_{2g}^3 e_g^0$). These differences must be fully considered in a more complete theoretical description for the spontaneous Hall effect.

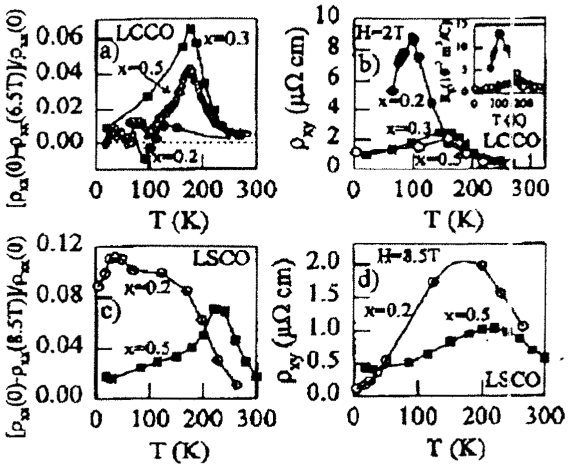

Fig. 1. Magnetoresistance and Hall resistivity of ferromagnetic $La_{1-x}Ca_xCoO_3$ and $La_{1-x}Sr_xCoO_3$.

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